

PATENT SPECIFICATION

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(54) THERMOPLASTIC MOLDING COMPOSITION

(71) We, GENERAL ELECTRIC COMPANY, a Corporation organized and existing under the laws of the State of New York, United States of America, of 1 River Road, Schenectady, 12305, State of New York, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention is directed to a novel thermoplastic molding composition comprising an aromatic carbonate polymer and a thermoplastic polyester resin derived from cyclohexanedimethanol. This composition may additionally contain a reinforcing amount of a reinforcing filler.

Polycarbonates are well known and have been widely employed for the production of thermoplastic molded articles.

Polycarbonate is a high-performance plastic with good impact strength. In addition to ductility (impact strength), general-purpose polycarbonate has high transparency, wide temperature limits (high impact resistance below -60°C and a UL thermal endurance rating of 115°C with impact), good dimensional stability high creep resistance and electrical properties which qualify it as sole support for current carrying parts (up to 125°C without loss of impact strength).

Polycarbonate has low water absorption, good stain resistance and a wide range of colorability. A weak area for polycarbonate is its relatively limited range of chemical resistance, which necessitates careful appraisal of applications involving contact with certain organic solvents, some detergents, strong alkali, certain fats, oils, and greases. Also, another weak area of polycarbonates is that they have high melt viscosities which makes them to some extent difficult to mold.

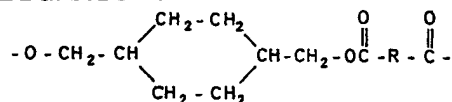
Attempts have been made to blend polycarbonates with various polymeric systems. Generally, the polycarbonate is not miscible with uniformity with the other polymer system. However, U.S. Patent 3,218,372, for example, describes compositions of polyalkylene terephthalate and polycarbonate resins. These unreinforced compositions are described as having a reduced melt viscosity and as having a higher ductility than the polyalkylene terephthalate resins. Even in the occasional instances where the polycarbonate is miscible with the other polymer system, the properties which make polycarbonate a high performance plastic are generally reduced without additional advantages.

It has been discovered that when an aromatic carbonate polymer is added to a polyester resin derived from cyclohexane-dimethanol, the polycarbonate is compatible with this polyester over a wide range. An article molded from this composition retains the transparent characteristics of an article molded from the polycarbonate. Also, this is achieved without any appreciable decrease in the mechanical and physical properties, such as Gardner impact strength, tensile strength or flexural strength, of a typical polycarbonate molding composition. Also, the composition of the present invention has a reduced melt viscosity which results in easier molding. Further, an article molded from this composition has better hydrolysis resistance and reduced water vapor transmission rate (this is important in bottles and packaging).

The present invention provides a thermoplastic molding composition which comprises a compatible blend of

(a) an aromatic carbonate polymer; and

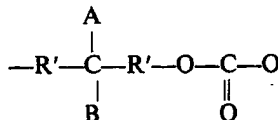
(b) a polyester resin containing recurring units of the formula:



wherein the substituted cyclohexane ring is the *cis*- or *trans*- isomer or a mixture thereof, and R represents an organic radical containing from 6 to 20 carbon atoms which is the decarboxylated residue derived from a hexacarboxylic dicarboxylic acid.

In a preferred embodiment, the thermolastic composition of the invention also comprises a reinforcing amount of a reinforcing filler.

The aromatic carbonate polymer of the present invention generally has recurring units of the formula:



wherein each $-\text{R}'-$ is phenylene, halo-substituted phenylene or alkyl-substituted phenylene; and A and B are each hydrogen, hydrocarbon radicals free from aliphatic unsaturation or radicals which together with the adjoining

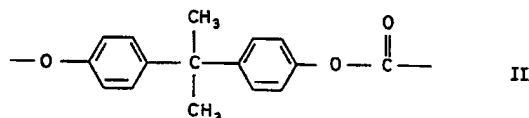


atom form a cycloalkane radical, the total number of carbon atoms in A and B being up to 12.

The aromatic carbonate polymer used according to this invention may be prepared by methods well known in the art and as described in U.S. Patent 3,989,672.

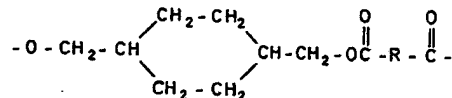
Also, included herein are branched polycarbonates wherein a polyfunctional aromatic compound is reacted with the dihydric phenol and carbonate precursor to provide a thermoplastic randomly branched polycarbonate wherein the recurring units of formula I. contain branching groups.

The preferred polycarbonate resins may be derived from the reaction of bisphenol-A and phosgene. These polycarbonates have from 10-400 recurring units of the formula:



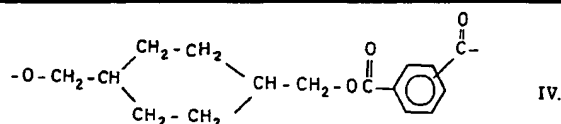
The polycarbonate should have an intrinsic viscosity between 0.3 and 1.0, preferably from 0.40 to 0.65 as measured at 25°C in methylene chloride.

The polyesters are prepared by condensing either the *cis*- or *trans*- isomer (or a mixture thereof) of 1,4-cyclohexanedimethanol with a hexacarboxylic dicarboxylic acid so as to produce a polyester having recurring units having the following formula:



wherein the substituted cyclohexane ring is the *cis*- or *trans*- isomer or a mixture thereof, and R represents an organic radical containing from 6 to 20 carbon atoms which is the decarboxylated residue derived from a hexacarboxylic dicarboxylic acid.

The preferred polyester resins may be derived from the reaction of either the *cis*- or *trans*- isomer (or a mixture thereof) of 1,4-cyclohexanedimethanol with a mixture of iso- and terephthalic acids. These polyesters have recurring units of the formula:



These polyesters can be produced by well known methods in the art such as those set forth in U.S. Patent 2,901,466.

Of course, it is understood that the polyester resins of this invention can be prepared by condensing 1,4-cyclohexanedimethanol and minor amounts of other bifunctional glycols with the hexacarboxylic dicarboxylic acid. These other bifunctional glycols include the polymethylene glycols containing from 2 to 10 or more carbon atoms such as ethylene glycol or butylene glycol.

Examples of hexacarboxylic dicarboxylic acids wherein the carboxy radicals are attached in para relationship to a hexacarboxylic residue indicated by R in formula III include terephthalic acid, *trans*-hexahydroterephthalic acid, *p,p'*-sulfonyldibenzoic acid, 4,4'-diphenic acid, 4,4'-benzophenonedicarboxylic acid, 1,2-di(*p*-carboxyphenyl) ethane, 1,2-di(*p*-carboxyphenoxy) ethane, and 4,4'-dicarboxydiphenyl ether, and mixtures of these. All of these acids contain at least one hexacarboxylic nucleus. Fused rings can also be present such as in 1,4- or 1,5-naphthalenedicarboxylic acid. The hexacarboxylic dicarboxylic acids are preferably those containing a *trans*-cyclohexane nucleus or an aromatic nucleus containing from one to two benzene rings of which at least one has the usual benzeneoid unsaturation. Of course, either fused or attached rings can be present. All of the compounds named in this paragraph come within the scope of this preferred group. The preferred dicarboxylic acid is terephthalic acid, or mixtures of terephthalic and isophthalic acids.

These polyesters should have an intrinsic viscosity between 0.40 and 2.0 dl./g. measured in a mixture of 40% tetrachloroethane/60% phenol solution or a similar solvent at 25°C—30°C. Especially preferred polyester will have an intrinsic viscosity in the range of 0.6 and 1.2 dl./g.

The reinforcing agents may be selected from finely divided metals, e.g. aluminum, iron or nickel metal oxides and non-metals, such as carbon filaments, silicates, such as mica, aluminum silicate (clay), talc, asbestos, titanium dioxide, Wollastonite, novaculite, potassium titanate and titanate whiskers, glass flakes, glass beads and fibers, and polymeric fibers and combinations thereof.

Although it is only necessary to use a reinforcing amount of the reinforcing agent, from 1—60% by weight of the total weight of the composition may comprise the reinforcing agent. A preferred range is from 5—40% by weight.

The preferred reinforcing agents are of glass, and it is preferred to use fibrous glass filaments, mixtures of glass and talc, glass and mica and glass and aluminum silicate, for example. The preferred filaments for plastics reinforcement are made by mechanical pulling. Preferably, the glass filament diameters range from 0.00012 to 0.00075 inch, but this is not critical to the present invention.

The aromatic carbonate polymer may comprise from 1 to 99% by weight of the composition and the polyester resin derived from cyclohexanedimethanol resin may correspondingly comprise from 99 to 1% by weight of the composition. Preferably, the aromatic carbonate polymer comprises from 25 to 98% by weight of the composition and the polyester derived from cyclohexanedimethanol comprises from 2 to 75% by weight of the composition.

The present composition may be prepared by any standard procedure and the particular method employed is not critical. For example, pellets made from the polycarbonate resin can be blended with pellets made from the polyester resin in an extruder under standard conditions.

Obviously, other materials can also be employed with the composition of this invention and include such materials as antistatic agents, pigments, mold release agents, thermal stabilizers, impact modifiers, extenders, UV stabilizers, nucleating agents, and flame retardants.

Illustrative flame retardants are disclosed in U.S. Patents 3,917,559, 3,919,167, 3,926,908, 3,931,100, 3,933,734, 3,948,851, 3,951,910, 3,953,396 and 3,940,366.

Preferred flame retardant additives are metal salts of sulfonic acids. These are the alkali metal or alkaline earth metal salts of: monomeric or polymeric aromatic sulfonic acids; monomeric or polymeric aromatic sulfone sulfonic acids; aromatic ketone sulfonic acids; heterocyclic sulfonic acids; halogenated methane sulfonic acids; halogenated non-aromatic carboxylic acids; aromatic sulfide sulfonic acids; monomeric or polymeric aromatic ether sulfonic acids; aliphatic and olefinic

sulfonic acids; monomeric or polymeric phenol ester sulfonic acids; unsubstituted or halogenated oxocarbon acids; monomeric and polymeric aromatic carboxylic acids and esters and monomeric and polymeric aromatic amide sulfonic acids.

5 These flame retardant additives are used in minor amounts, preferably from 0.01 to 10 weight percent, based on the weight of the combination of the aromatic carbonate polymer and polyester. 5

10 In order to more fully and clearly illustrate the present invention, the following specific examples are presented. It is intended that the examples be considered as illustrative rather than limiting the invention disclosed and claimed herein. In the examples, all parts and percentages are on a weight basis unless otherwise specified. 10

EXAMPLE I

The following compositions were prepared by melt blending the components in an extruder (all parts are by weight):

15		A	B	C	D	E	F	15
	polycarbonate resin*	100	75	70	60	40	35	
	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	—	25	—	40	60	35	
	glass	—	—	30	—	—	30	

20 * Lexan 101 (General Electric Co. "Lexan" is a Registered Trade Mark) 20
** Kodar A150 (Eastman Kodak Co.)

The compositions of Example I were tested and found to have the following physical properties as set forth in Table I.

25		A	B	C	D	E	F	25
	Tensile strength (psi)	9,500	8,532	16,676	8,297	7,355	15,528	
	Tensile elongation (%)	110	148	5.8	158	196	6.0	
	Flexural strength (psi)	13,595	13,572	25,466	12,878	12,110	23,591	
	Flexural modulus (psi)	376,200	341,400	1,092,200	325,400	310,200	1,035,800	

30 **EXAMPLE II** 30
The following compositions were prepared by melt blending the components in an extruder (all parts by weight):

35		G	H	I	J	K	L	M	35
	polycarbonate resin*	100	75	70	60	50	40	35	
	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	—	25	—	40	50	60	35	
	glass	—	—	30	—	—	—	30	

* Lexan 101
** Kodar A150

40 The compositions of Example II were tested and found to have the following physical property as set forth in Table II. 40

45		G	H	I	J	K	L	M	45
	Gardner Impact Strength (in.-lbs.)	560	560	8	560	560	560	8	

45 **EXAMPLE III** 45
The following compositions were prepared by melt blending the components in an extruder (all parts by weight):

50		N	O	P	Q	R	S	T	U	50
	polycarbonate resin*	100	90	75	60	50	40	30	—	
	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	—	10	25	40	50	60	90	100	

* Lexan 101
** Kodar A150

The melt viscosity of the compositions of Example III were measured and the results are set forth in Table III.

TABLE III

	N	O	P	Q	R	S	T	U	
5 Melt Viscosity (poise)	9872	8710	6452	4970	4264	3551	2440	2303	5
Time (sec.)	43.3	38.2	28.3	21.8	18.7	15.6	10.7	10.1	

EXAMPLE IV

The following compositions were prepared by melt blending the components in an extruder (parts by weight):

10		V	W	10
	polycarbonate resin*	100	95	
	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	—	5	
15	* Lexan 101 ** Kodar A150			15

The compositions of Example IV were tested and found to have the following physical properties as set forth in Table IV.

TABLE IV

20	Melt Viscosity (poise)	V	W	20
	Notched izod impact strength (ft.-lbs./in.)	3250	2485	
	Double gated izod impact strength (ft.-lbs)	14.7	15.5	
		40	40	

EXAMPLE V

The following compositions were prepared by melt blending the components in an extruder (parts by weight):

25		X	Y	25
	polycarbonate resin*	91	87.6	
	glass	0	4	
30	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	9	8.4	30
	* Lexan 101 ** Kodar A150			

The compositions of Example V were tested and found to have the following physical properties as set forth in Table V.

TABLE V

35		X	Y	35
	Notched izod impact strength (ft.-lbs/in.)	1.9	3.3	
	Flexural modulus (psi)	500,000	496,000	
	Flexural strength (psi)	15,000	15,400	

EXAMPLE VI

The following compositions were prepared by melt blending the components in an extruder (parts by weight):

40		A'	B'	40
	polycarbonate resin*	75	75	
45	poly(1,4-cyclohexanedimethanol terephthalate-co-isophthalate)**	25	—	45
	poly(ethylene terephthalate)***	—	25	
50	* Lexan 101 ** Kodar A150 *** Vitel VFR 3801 ("Vitel" is a Registered Trade Mark)			50

The compositions of Example VI were tested and found to have the following physical properties as set forth in Table VI.

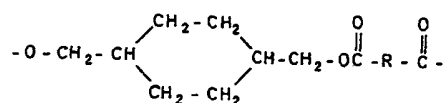
TABLE VI

		A'	B'	
5	Light Transmission (%)	87	35	5
	Tensile elongation (%)	148	108	

From the preceding Tables, it can be seen that the composition of the instant invention retains the useful properties of aromatic polycarbonate resins while additionally demonstrating improved melt flow properties without loss of impact strength.

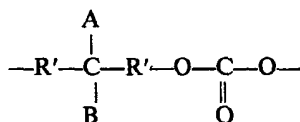
WHAT WE CLAIM IS:—

1. A thermoplastic molding composition which comprises a compatible blend of
 - (a) an aromatic carbonate polymer; and
 - (b) a polyester resin containing recurring units of the formula:



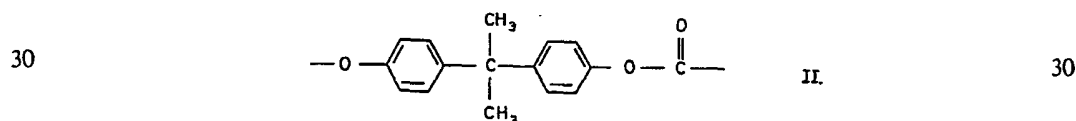
wherein the substituted cyclohexane ring is the *cis*- or *trans*- isomer or a mixture thereof, and R represents an organic radical containing from 6 to 20 carbon atoms which is the decarboxylated residue derived from a hexacarboxylic acid.

2. A composition as claimed in Claim 1, wherein the polycarbonate has recurring units of the formula:

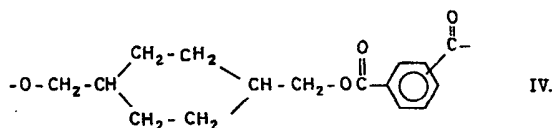


wherein each —R'— is phenylene, halo-substituted phenylene or alkyl-substituted phenylene, and A and B are each hydrogen or hydrocarbon radicals free from aliphatic unsaturation, or together with the adjacent carbon atom form a cycloalkylene radical, the total number of carbon atoms in A and B being up to 12.

3. A composition as claimed in Claim 2, wherein the polycarbonate has the recurring unit



4. A composition as claimed in any preceding claim, wherein the polyester resin contains recurring units of the formula:



5. A composition as claimed in Claim 4, wherein the polyester resin is derived from either the *cis*- or *trans*- isomer (or a mixture thereof) of 1,4-cyclohexanedimethanol with a mixture of iso- and terephthalic acids.

6. A composition as claimed in any preceding claim, which comprises:
 - (a) from 25 to 98% by weight of the aromatic carbonate polymer; and correspondingly
 - (b) from 2 to 75% by weight of the polyester resin.

7. A composition as claimed in Claim 6 which comprises from 1 to 60% by weight of a reinforcing filler.
8. A composition as claimed in Claim 7, wherein the reinforcing filler is glass filler.
- 5 9. A composition as claimed in Claim 7, wherein the reinforcing filler is glass and mica. 5
10. A composition as claimed in Claim 7, wherein the reinforcing filler is glass and talc.
- 10 11. A composition as claimed in Claim 1, substantially as hereinbefore described with reference to any of the Examples. 10

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